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Opportunities for nuclear materials studies using APS high-energy synchrotron X-rays

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ATR National Scientific User Facility User's Week

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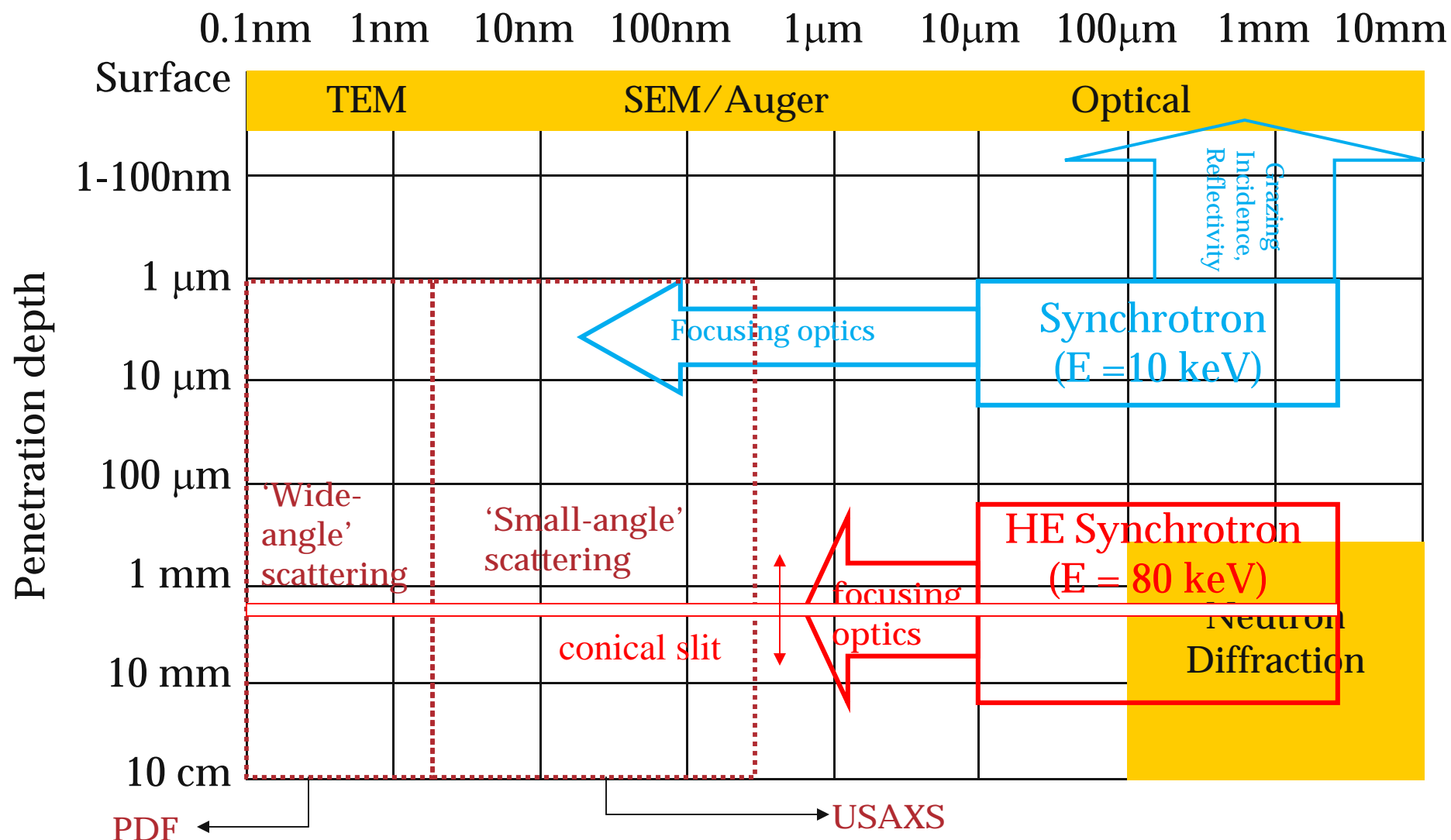
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Outline

- Survey of probe sizes & techniques
- Sector 1-ID beamline for high-energy x-ray studies
- Case study
 - In-situ evaluation of hydrides in Zr-alloys
- Summary
- Outlook

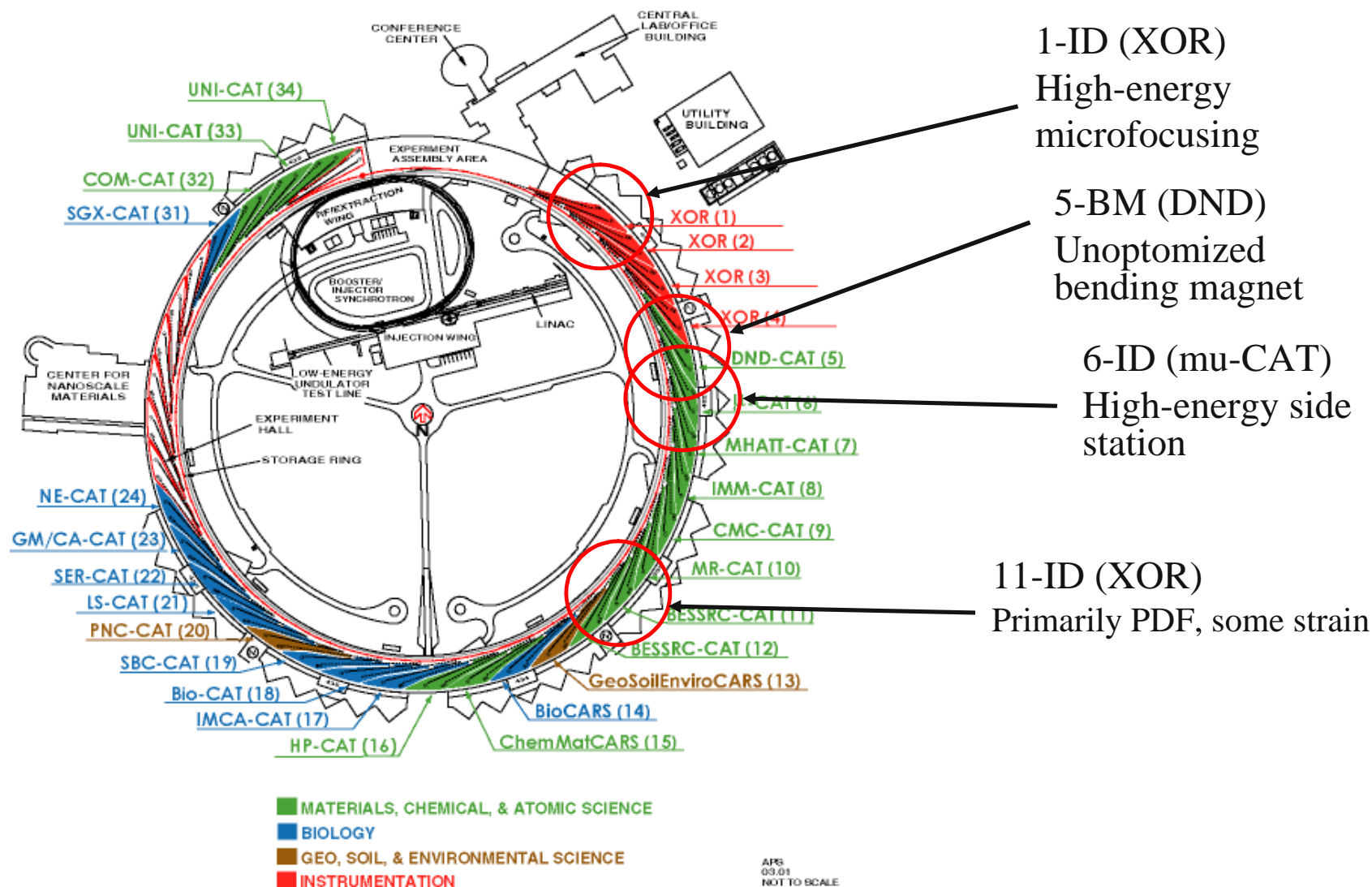
Probe sizes of selected techniques

Spatial resolution (1-d or 2-d)



High-energy x-ray studies at the APS

APS Collaborative Access Teams by Sector & Discipline



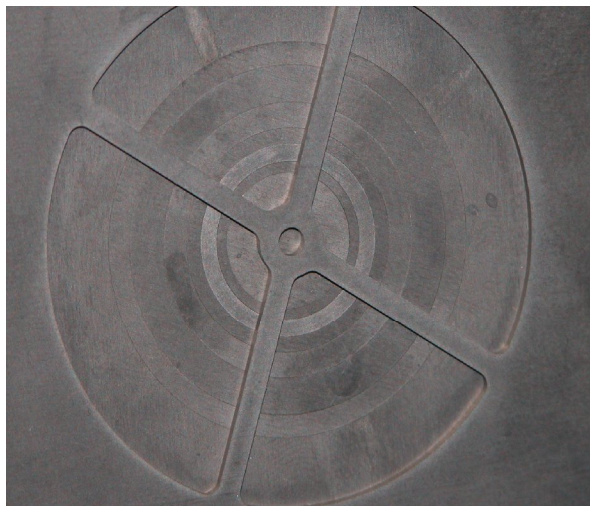
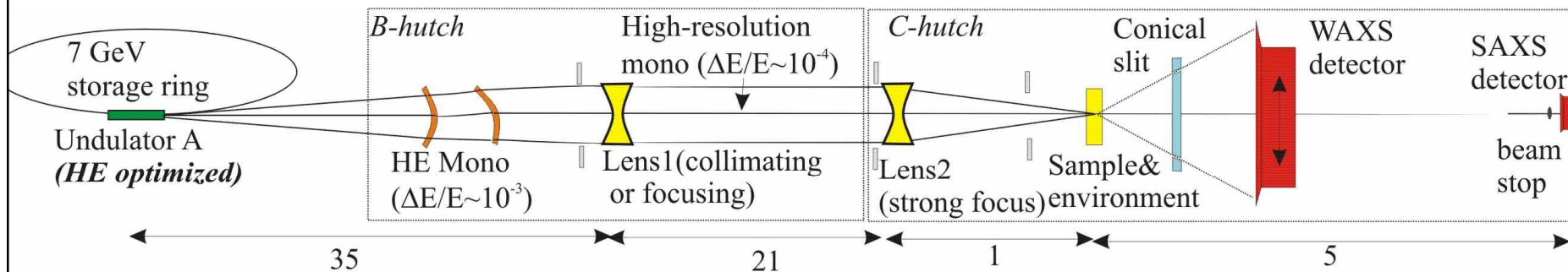
High-energy scattering at 1-ID

1-ID: simultaneous SAXS / WAXS measurements with high spatial (micron-level) and temporal (sub-second) resolution

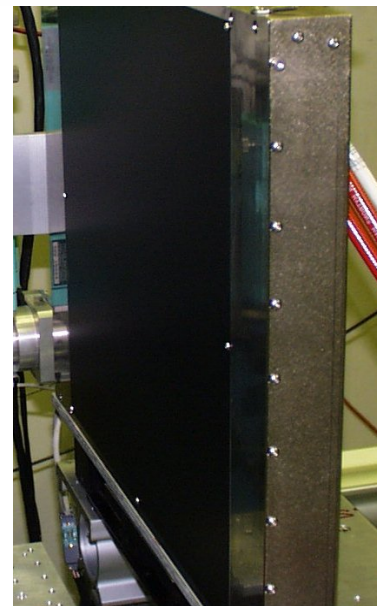
- WAXS: strain, texture, phase ID
- SAXS: porosity, nano-particle formation/precipitation
- Full-field imaging for selected applications
- Focus on *in situ* measurements
thermo-mechanical deformation, phase transformations, in-operando chemistry, processing operations, etc

- Typical energy = 80keV (bulk probe)
- Current exposure times $1 < t < 100$ sec
- Expected flux increases 10-100 X (via optimized undulator, improved optics, storage ring upgrade)
- Detectors should operate up to 100hz to account for increases

APS 1-ID beamline



Conical slit (7 rings)
• 3D WAXS spatial resolution

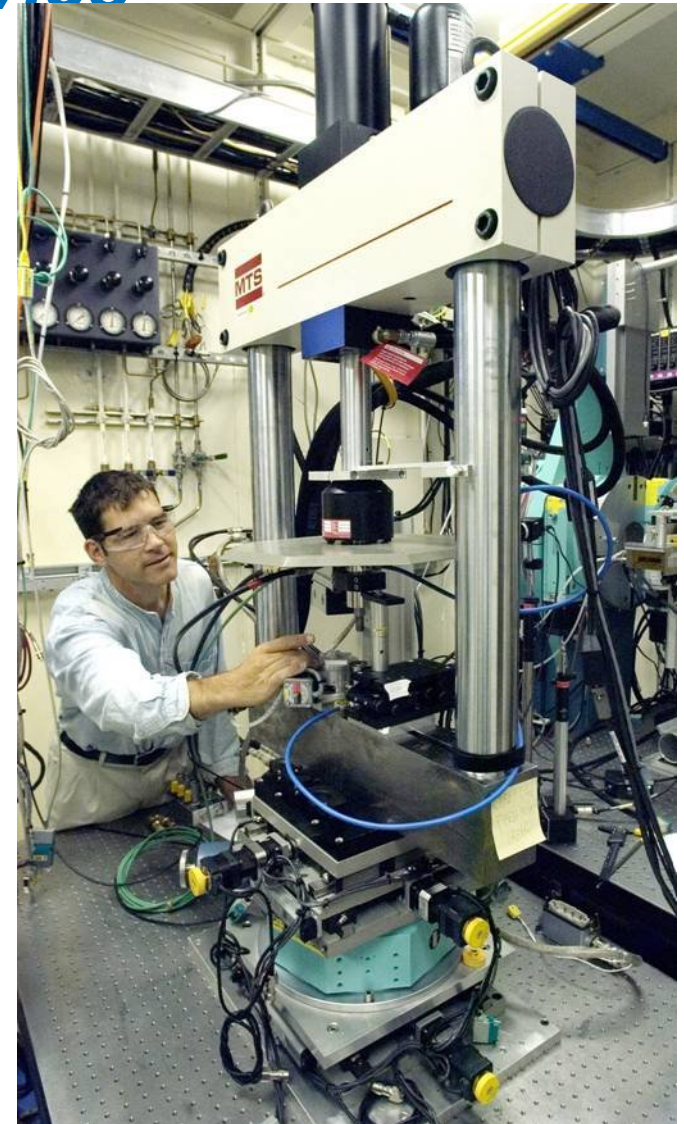


GE detector

In-situ thermo-mechanical device

Specification	Value
Loading mechanism	Hydraulics
Load rating	+/- 25 kN
Stroke	100 mm
Loading modes	Tension, bending, compression
Control modes	Load, displacement, strain
Max Temp @ applied load	~1200 C
Weight	200 kg
Degrees of freedom @ beamline	3 translations, 1 rotation, 2 tilts

Custom-designed by Materials Testing Systems (MTS)
Infrared furnace (Research Inc)



HE focusing using sawtooth refractive lenses

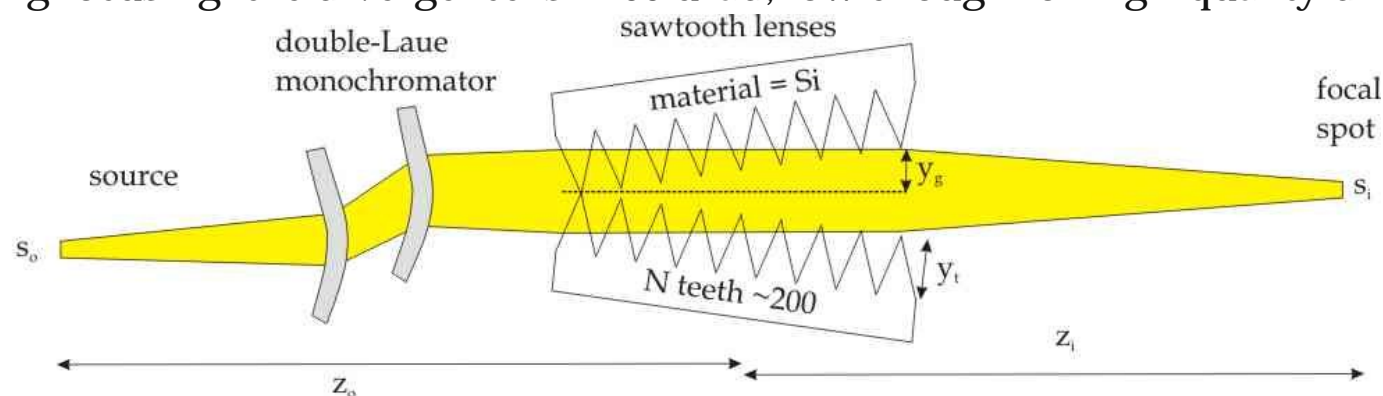
Collaborators: C. Ribbing (Uppsala) and B. Cederstrom (KTH), Sweden

Typical focal sizes:

18 μm (weak focusing; $z_o:z_i \sim 36:24 \text{ m}$)

1.5 μm (strong focusing; $z_o:z_i \sim 60:1 \text{ m}$)

Even at 'strong focusing' the divergence is $\sim 200 \text{ urad}$, low enough for high-quality diffraction



* high-energy monochromator is effectively brilliance preserving - minimal aberrations (some dependence on bend radii)

* use of pure material (single-crystal Si) gives minimal small-angle scattering from lens

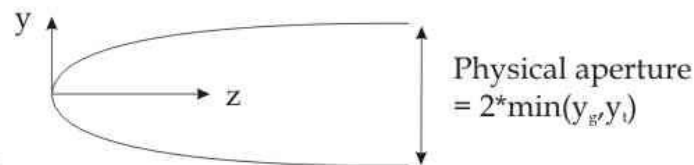
* no absorption along lens optic axis ($y=0$)

* lens focus distance z_i can be varied through y_s

* theoretical image size:

$$s_i = s_o(z_o/z_i)$$

canted sawtooth projection $y(z)$ is effectively parabolic (aberrations typically submicron)



Case study: hydrides in zirconium alloys

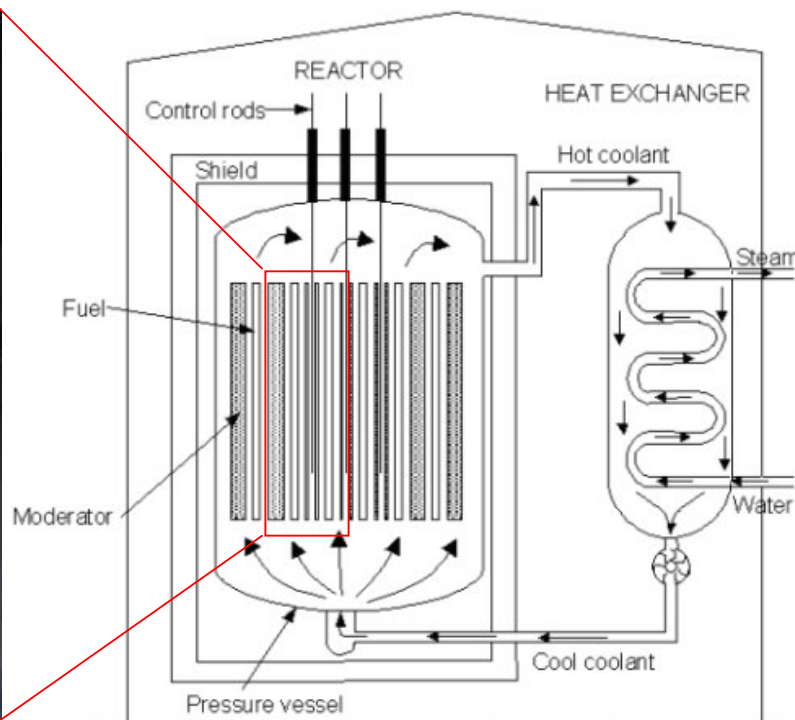
■ Hydrogen Embrittlement of Zirconium

■ Overview of Research Work

- *In-situ* deformation study of zirconium hydrides ✓
- Crack tip mapping in un-hydrided material ✓
- Crack tip mapping with previously grown hydrides ✓
- In situ growth of hydrides at crack tip (later this month)

Zr-hydrides: Background1

Reactors World Wide



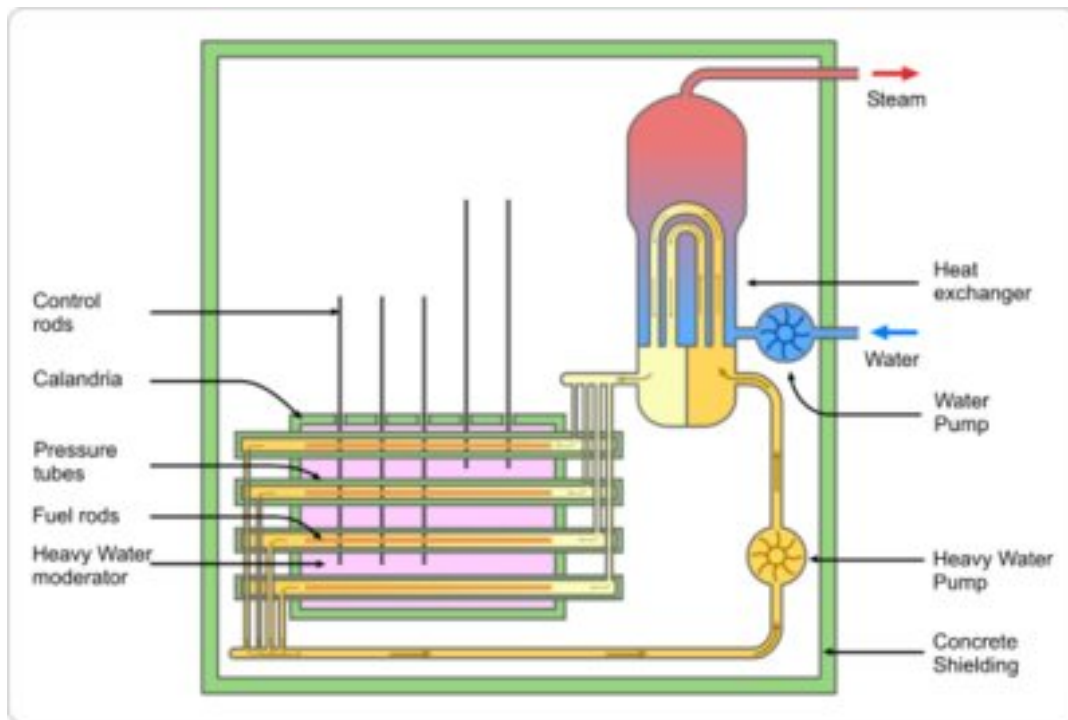
■ Zircaloy Fuel Cladding

- Pressurized or unpressurized H_2O coolant
- Temperatures range from 100 to greater than 300°C

Corrosion reaction at Zr surface: $\text{Zr} + 2\text{H}_2\text{O} \rightarrow \text{ZrO}_2 + 4\text{H}$

Zr-hydrides: Background2

CANDU Reactors

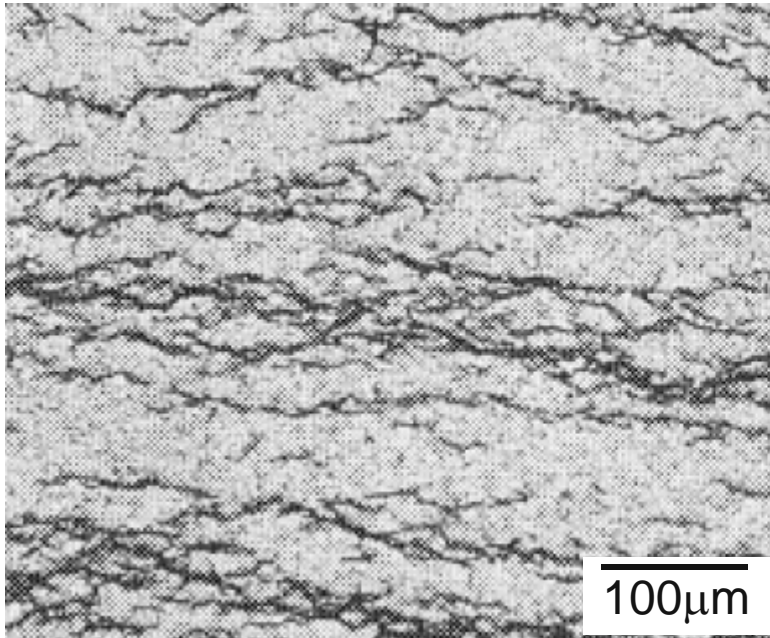


■ Zr-2.5Nb Pressure Tubes

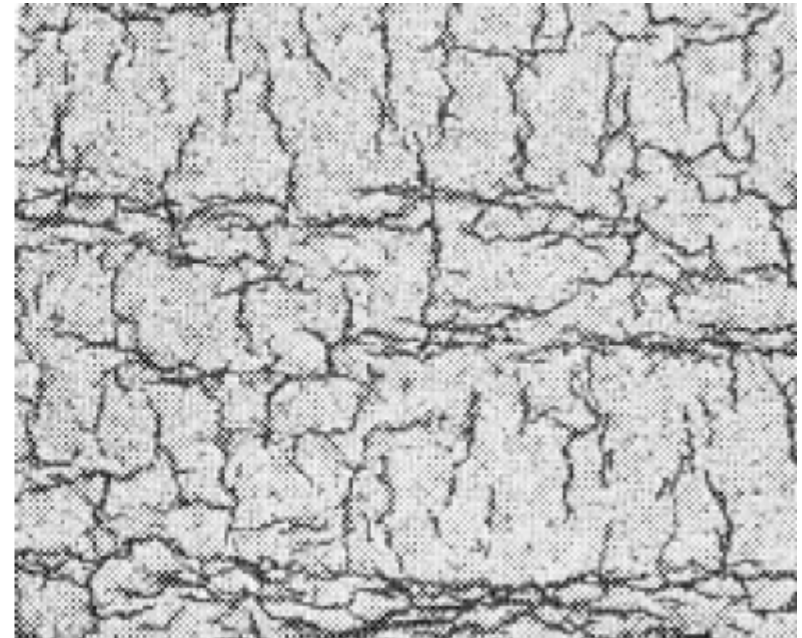
- D₂O coolant pressurized to ~ 10MPa
- Temperatures range from ~ 250°C (inlet) to 310°C (outlet)

Corrosion reaction at Zr surface: $\text{Zr} + 2\text{D}_2\text{O} \rightarrow \text{ZrO}_2 + 4\text{D}$

Hydride Characteristics



Cooled from above H Solubility Limit



Cooled from above H Solubility Limit

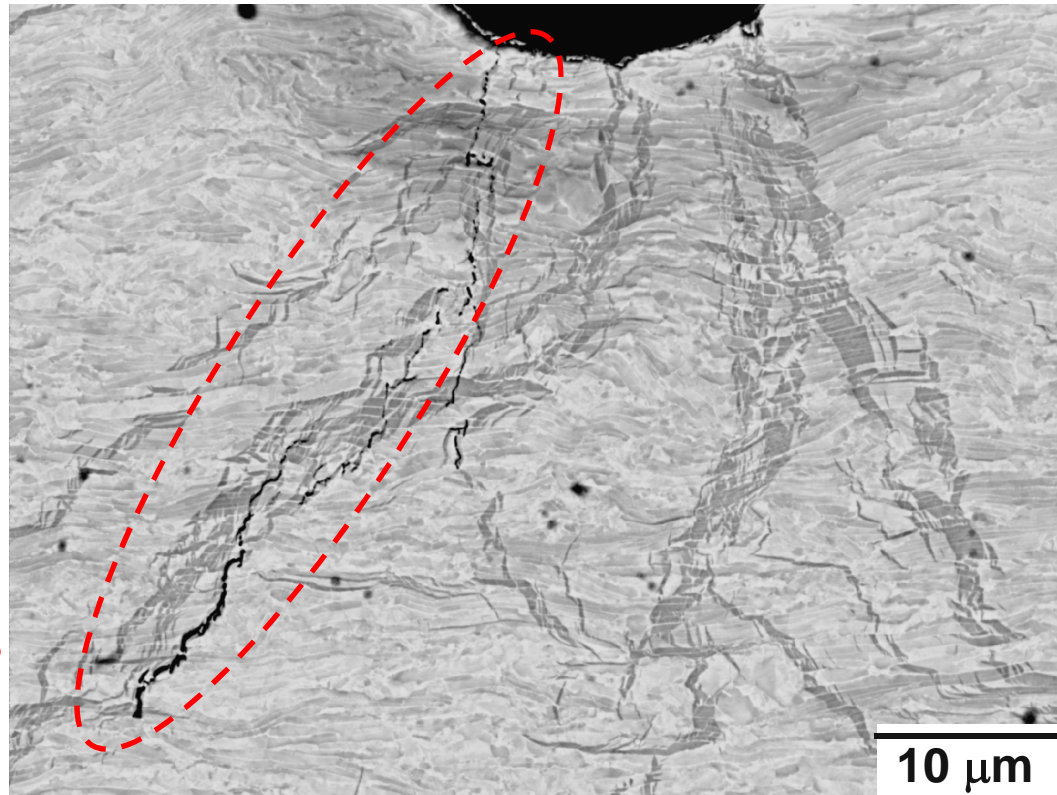
← **Tensile Stress** →

- Hydrides precipitate when solubility limit is exceeded
- Hydrides can reorient under an applied stress or in a temperature gradient or ...

DHC Characteristics

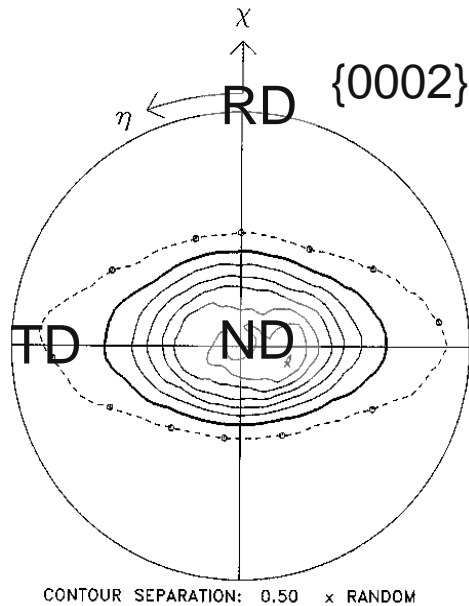
... in the stress field around a flaw or notch!

Cracked Hydrides



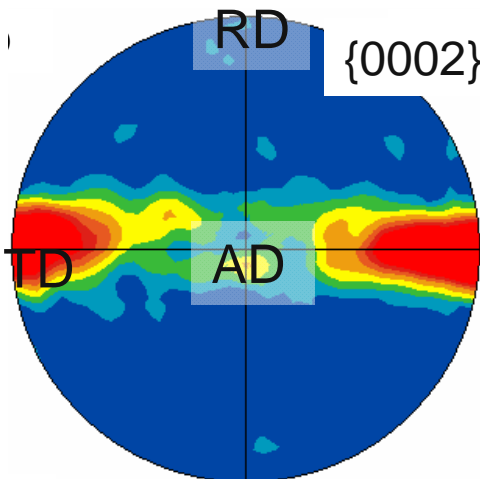
Failure of components occurs by the fracture and re-growth of crack tip hydrides - Delayed Hydride Cracking (DHC)

Source Materials



■ Rolled Zircaloy-2 Plate

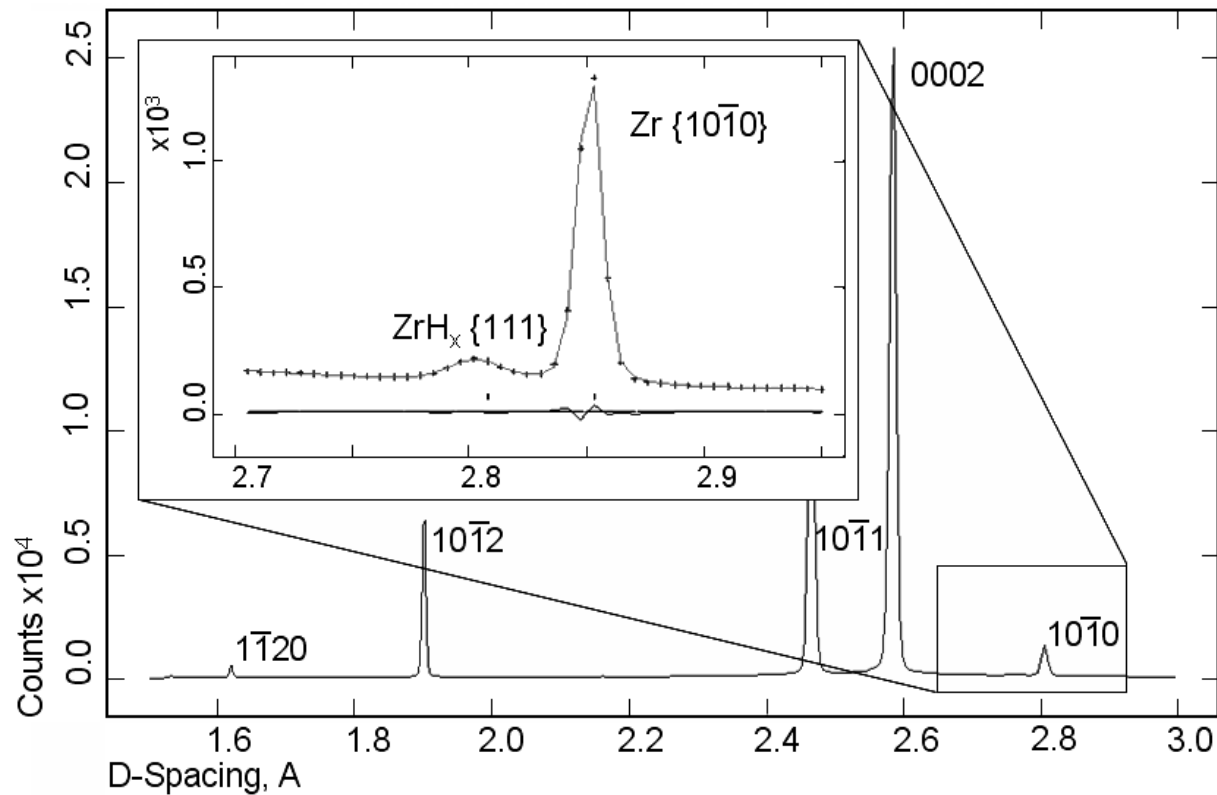
- Only α -phase Zr present
 - *Simplified diffraction pattern*
 - *Well characterized in terms of texture and mechanical properties*
- Initial hydride orientation determined by texture (typical of fuel sheathing)



■ Zr-2.5Nb Pressure Tube

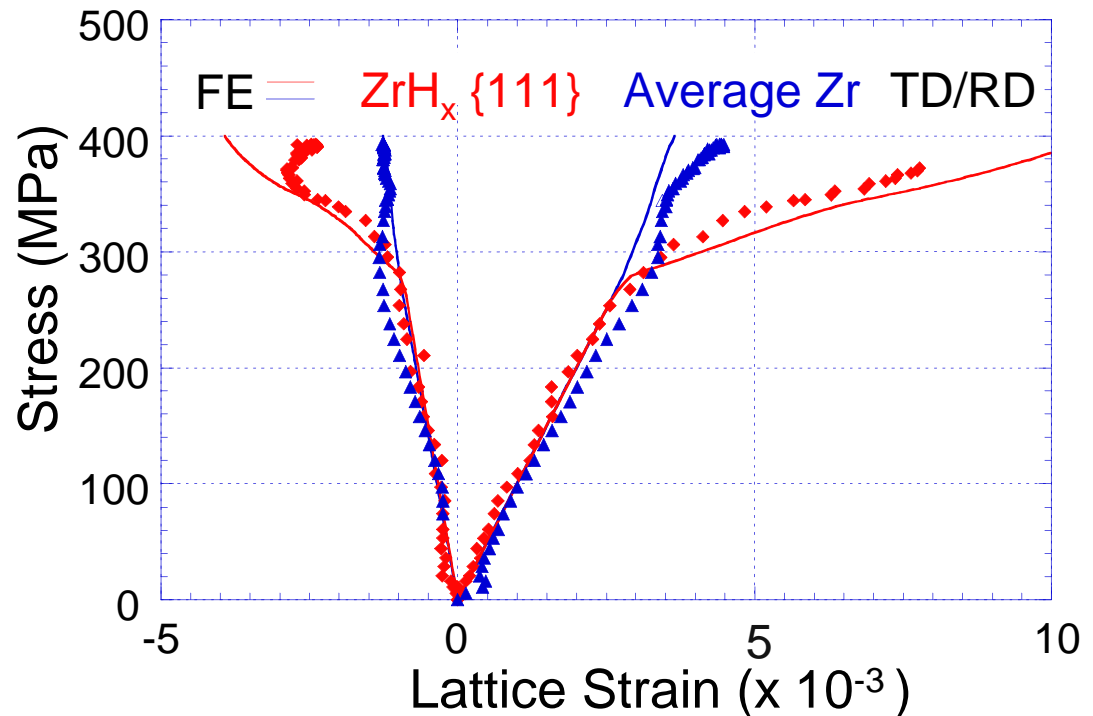
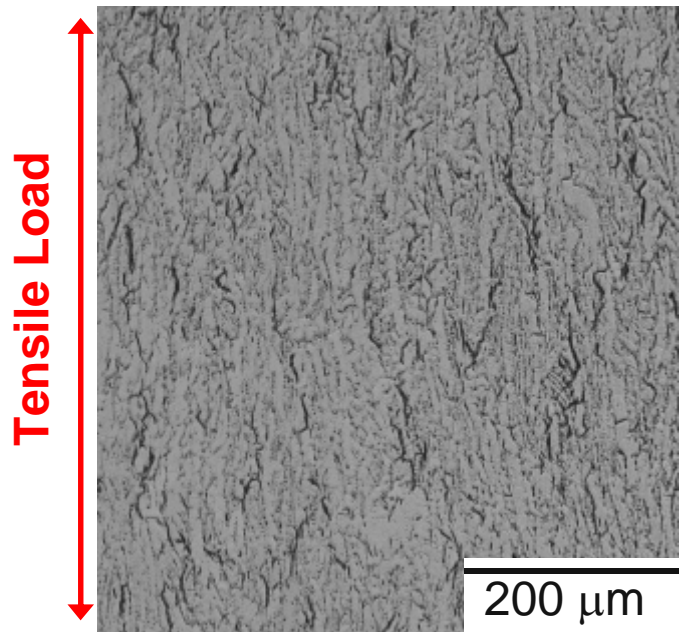
- α/β -phase Zr present
- Initial hydride orientation determined by texture and residual stress state

Hydride Diffraction Pattern



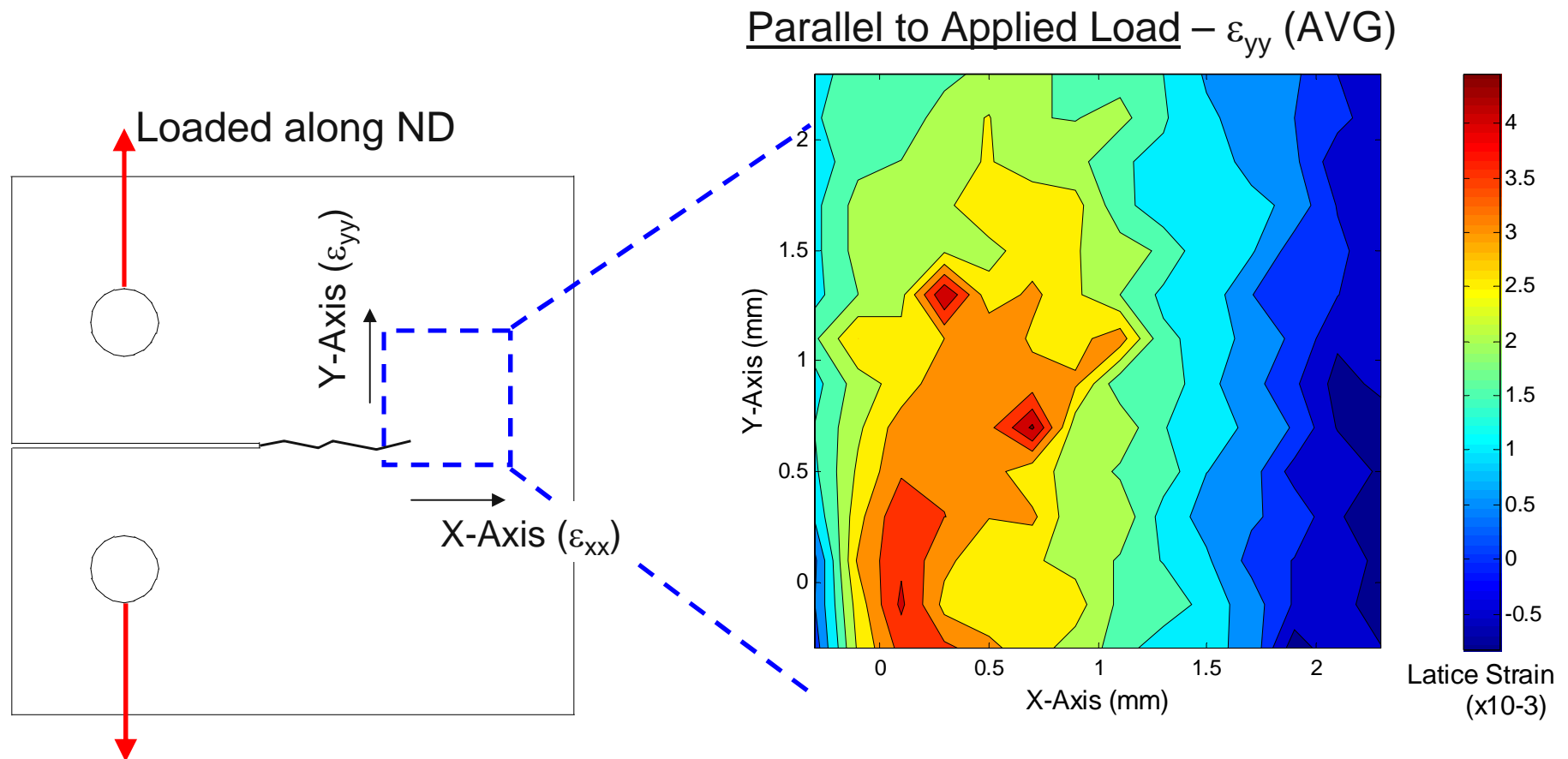
- Single peak fits (GSAS and Matlab)
- Diffraction directly measures the elastic strain in the lattice
 - Plastic behavior only inferred through load transfer behavior
- For comparison to elastic strain in Finite Element (FE) calculations, a weighted average of single diffraction peaks was used (multiplicity, texture, etc)

Overview: In-situ Bulk Deformation



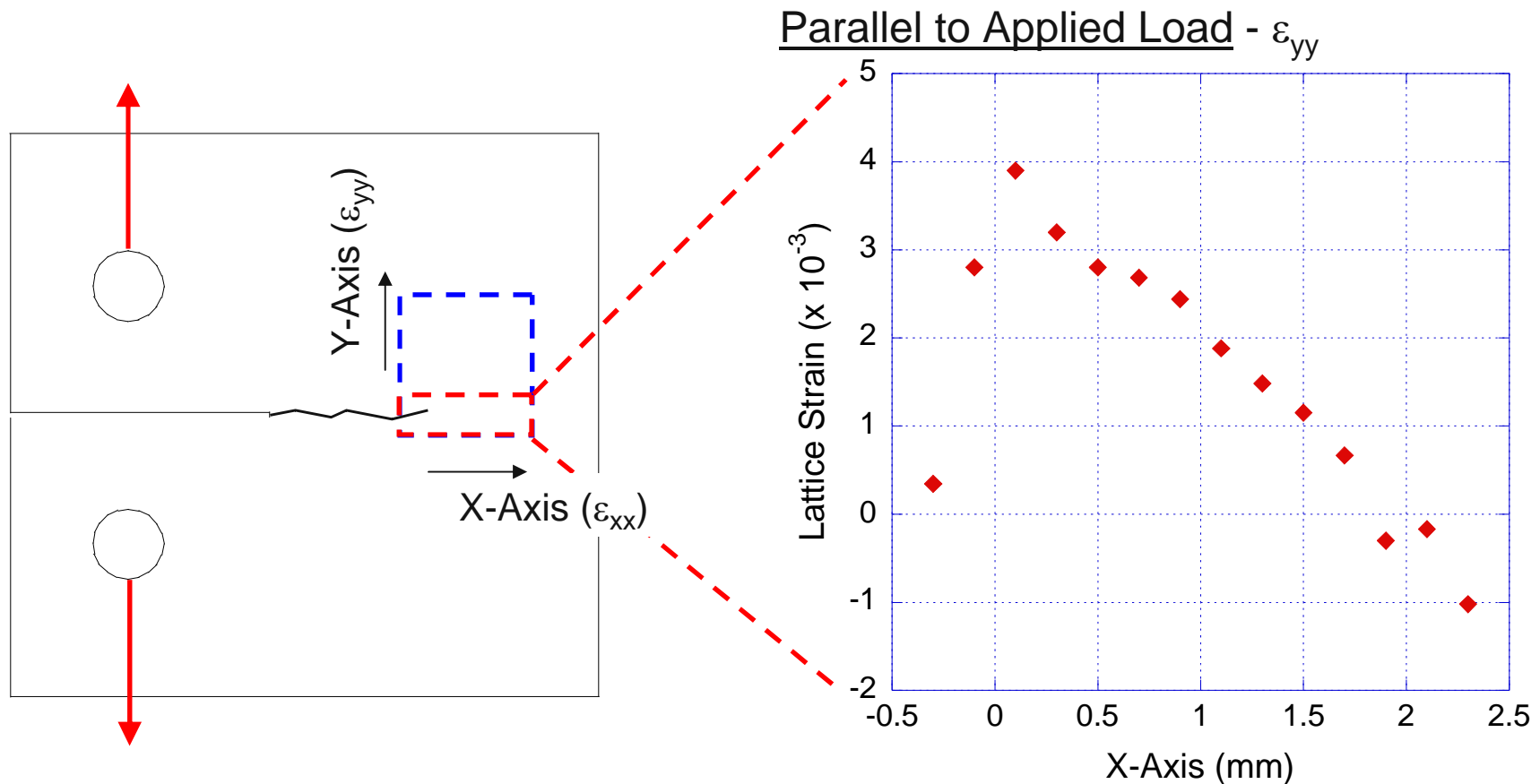
- X-ray diffraction allows measurement of hydride mechanical properties in-situ
- Finite Element Analysis (composite unit cell) captures load transfer to the hydride phase
- Short Fiber Composite Mechanics predicts critical size for load transfer and hydride fracture

Overview: Strain Mapping in Zircaloy-2



- $60\mu\text{m}^2$ beam size, with a diffraction pattern acquired every $200\mu\text{m}$
- Single Peak fits give strain & intensity.
 - Average strain calculated using a texture weighted average method

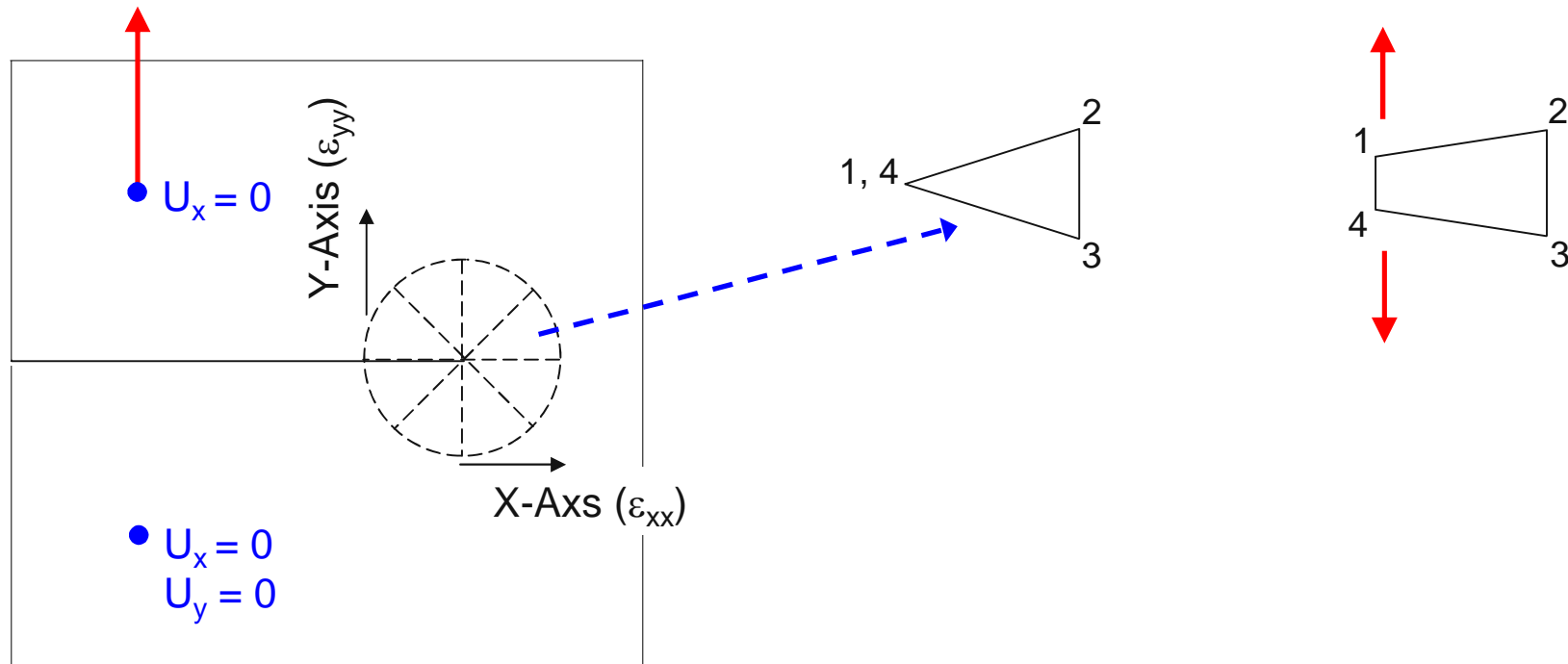
Overview: Strain Mapping in Zircaloy-2



- Take average within region -0.2, 0 and 0.2mm from nominal crack line to construct strain vs position plots
- Data compared to FE model

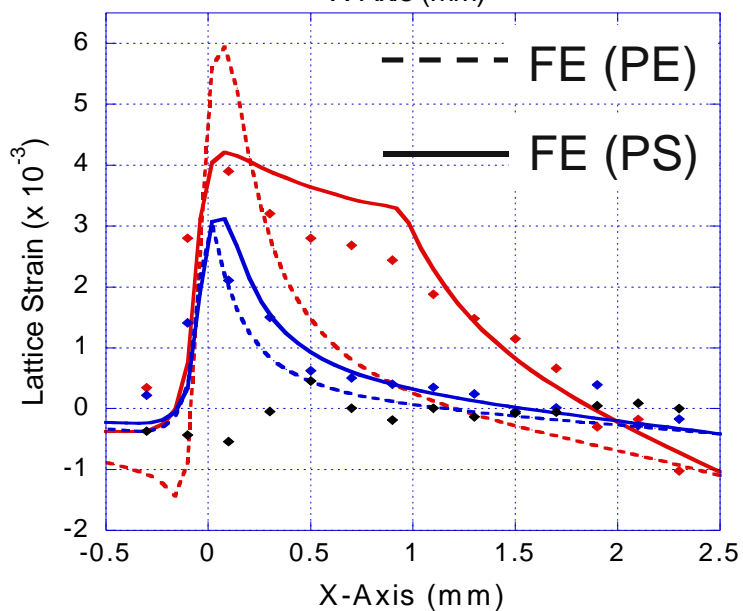
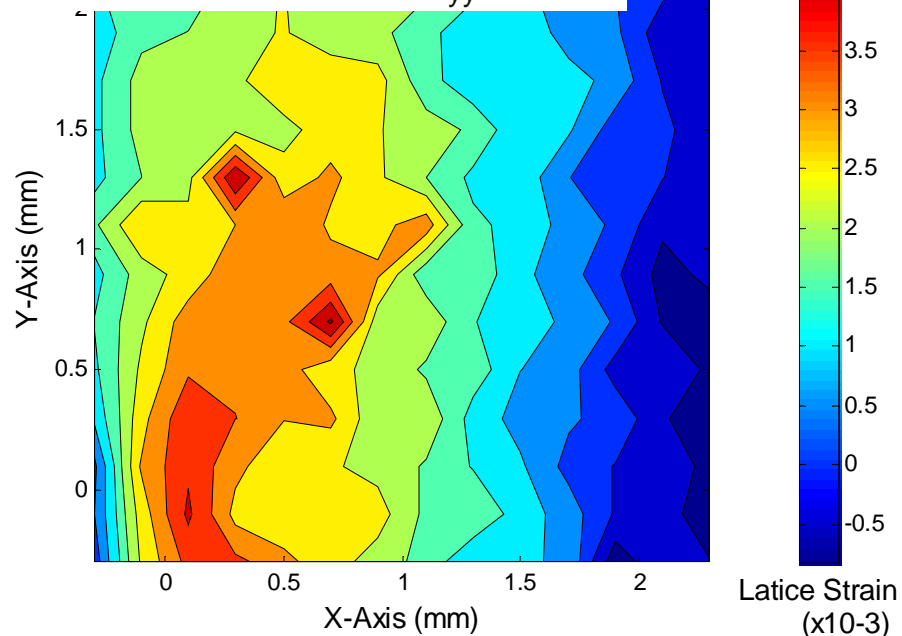
Overview: Strain Mapping in Zircaloy-2

Rosette used to mesh crack tip

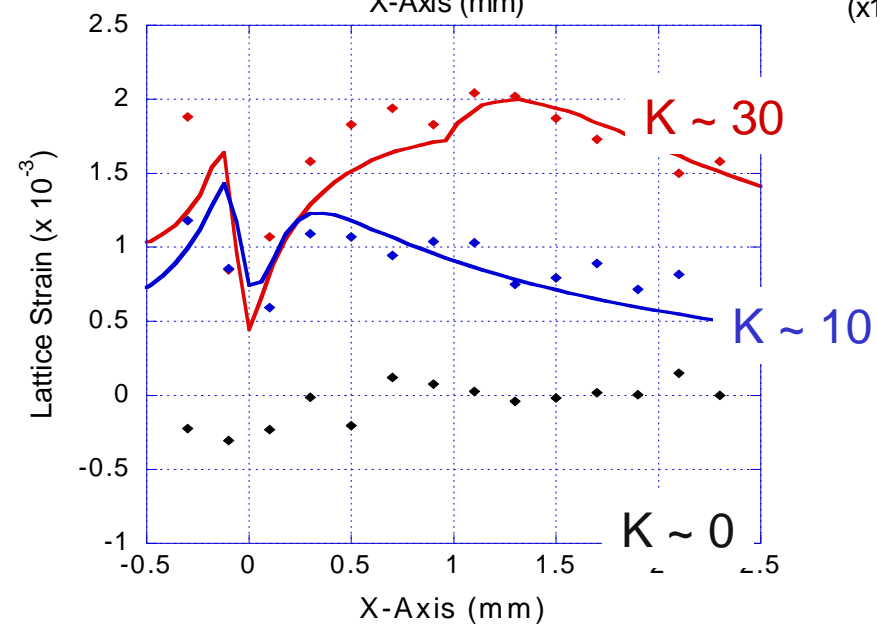
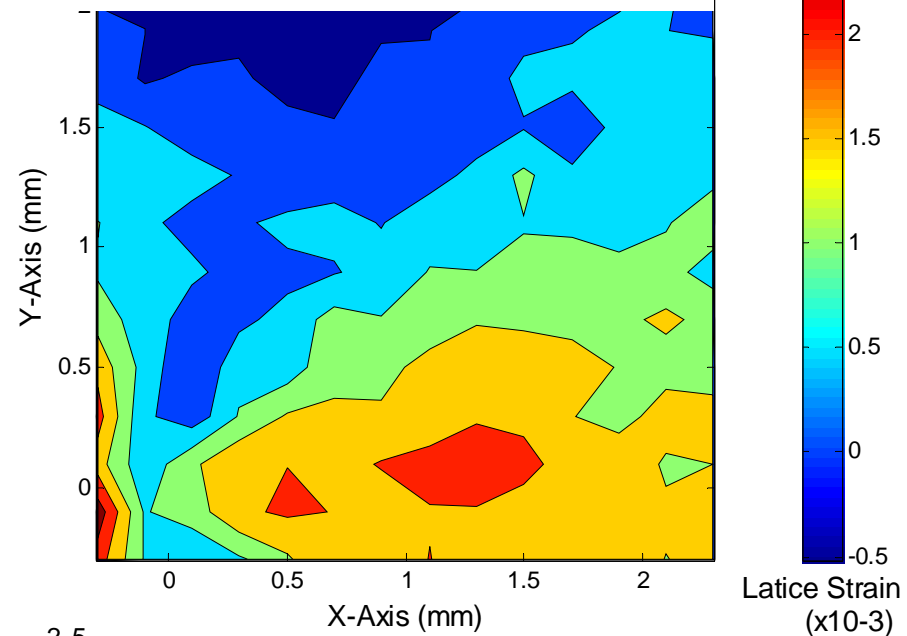


- 2D Plane Stress or Strain elements
- Rosette used at crack tip, quadrilateral elements collapsed to triangles
- Hill parameters used to account for plastic anisotropy
- Abaqus finite element package used

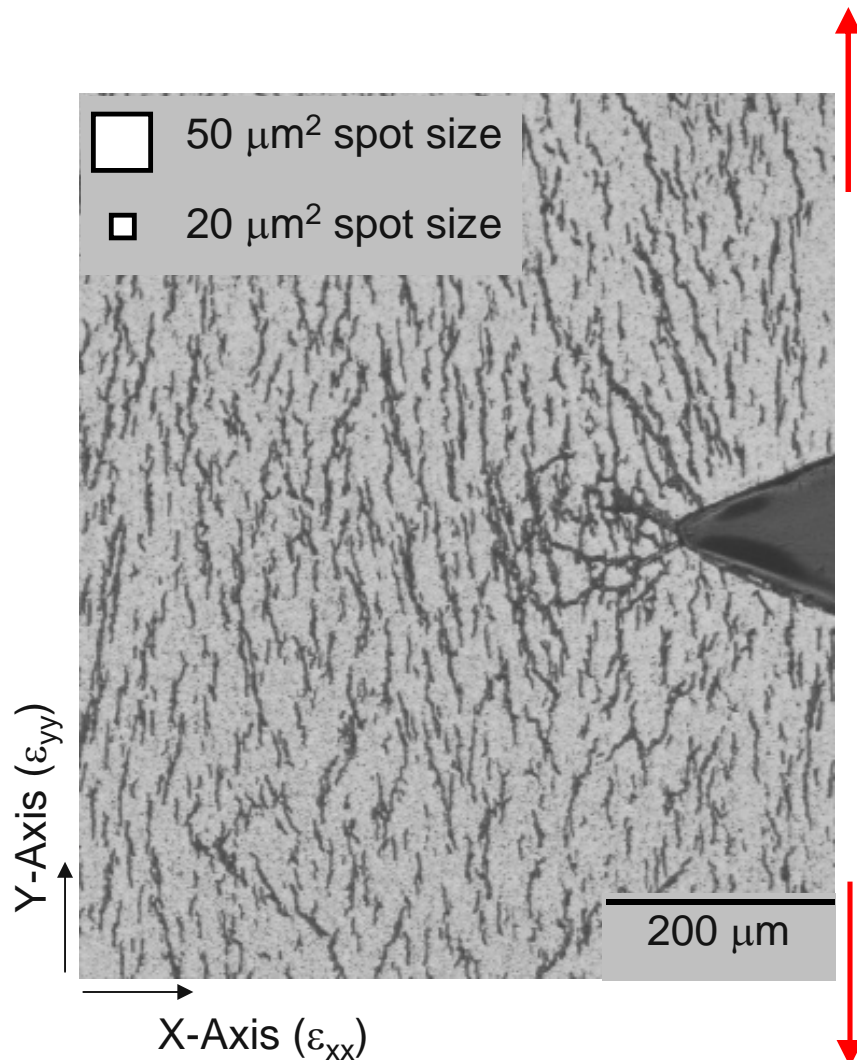
ND Parallel to Load – ε_{yy} (AVG)



ND Perpendicular to Load – ε_{xx} (AVG)



Overview: Hydride Strain Mapping



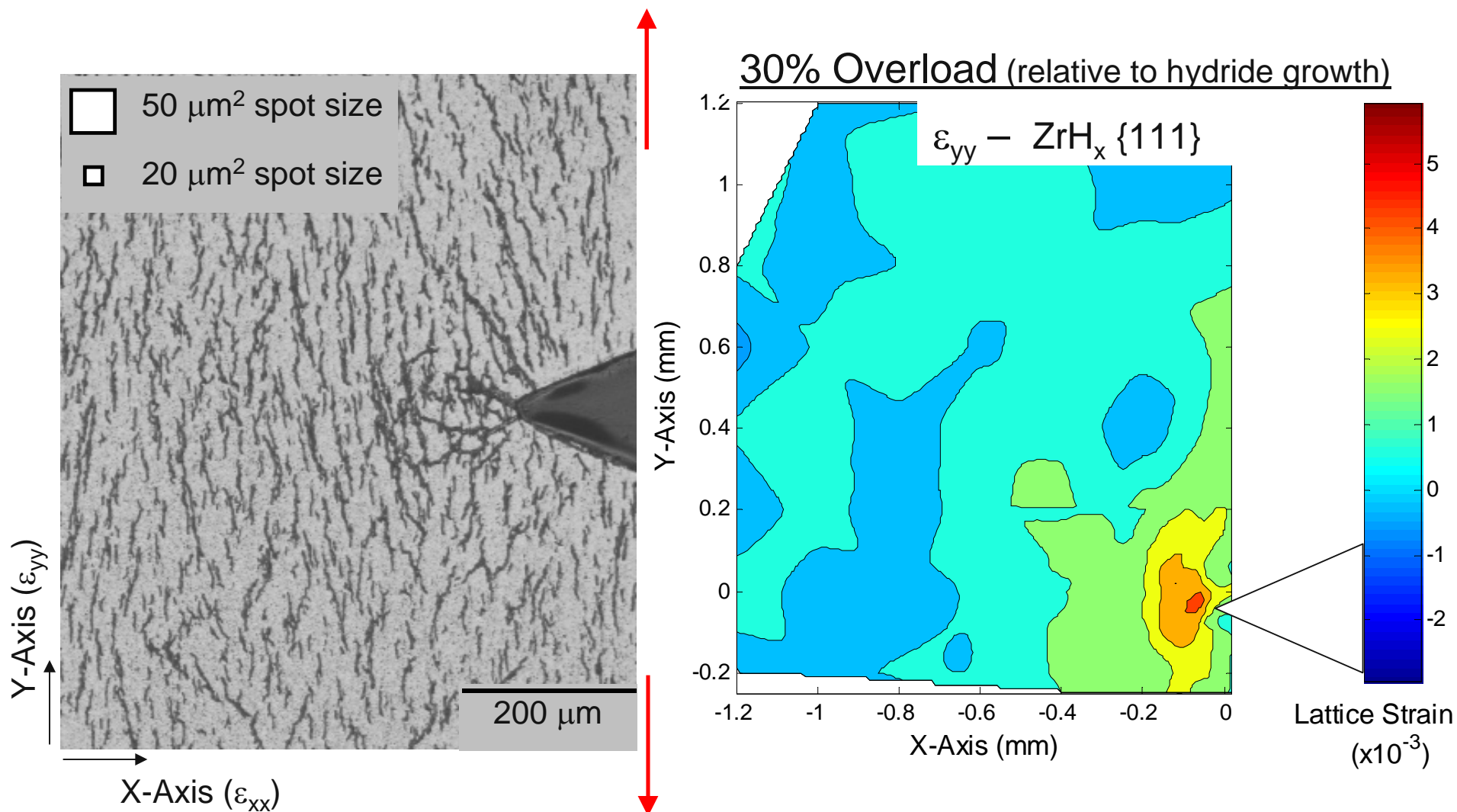
■ Zr-2.5Nb Pressure Tube

- 15 μm root radius notch
- Large $\sim 100 \mu\text{m}$ hydrides grown at notch prior to experiment

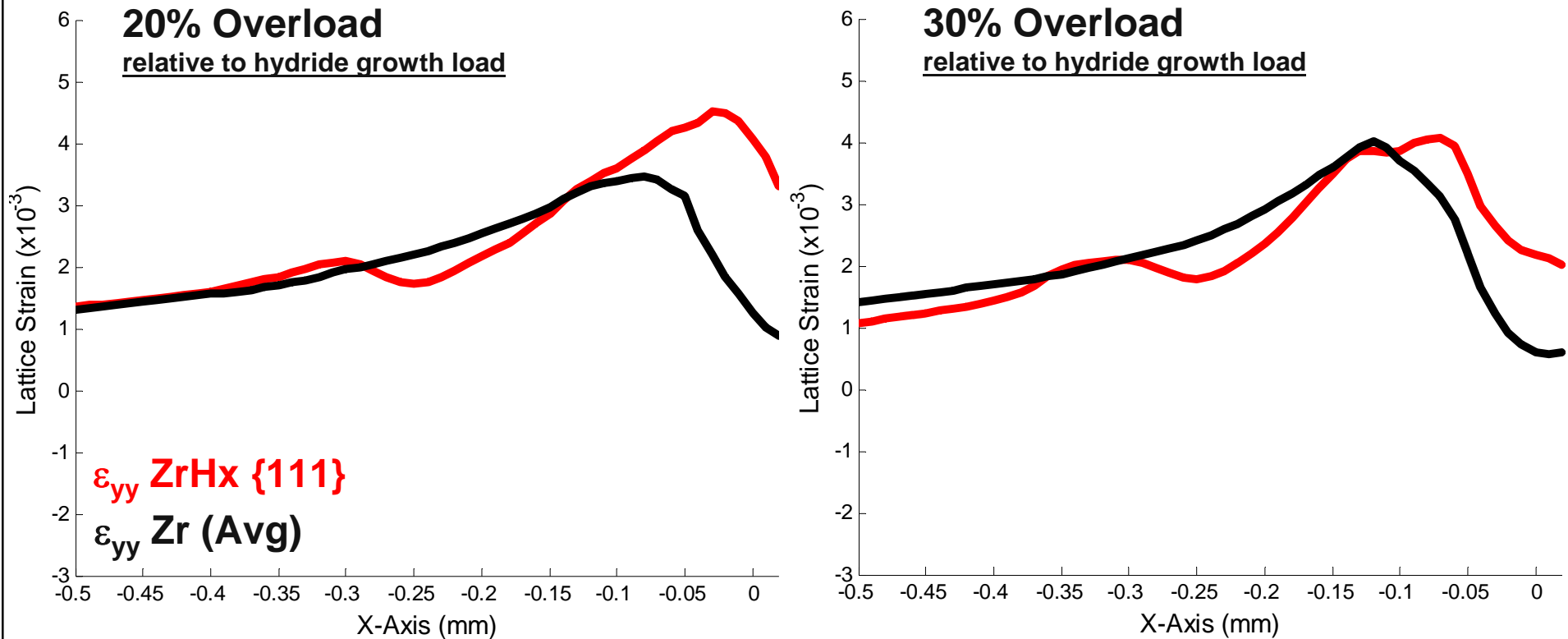
■ Loaded in-situ at 1-ID

- Incrementally loaded
- Mapped with 20 and 50 mm^2 spot size
- Tests at room temp and 250°C

Overview: Hydride Strain Mapping



Hydride Fracture



- Preliminary analysis indicates that hydride fracture can be resolved
- At a 20% overload, hydride is intact at the notch
- At a 30% overload, the notch tip hydride has fractured transferring load to the surrounding matrix

Summary

- High-energy x-ray scattering is a powerful technique for in-situ investigation of nuclear materials
- Sector 1 instrument benefits from:
 - 7 GeV synchrotron + undulator source
 - Optics:
 - *Brilliance-preserving monochromator*
 - *Refractive focusing lenses*
 - *Conical slit*
 - Two-dimensional detectors (fast and large)
- Hydride studies
 - High-intensity synchrotron permits detection of weak hydride peaks
 - Strain in each crystalline phase can be measured in-situ
 - Finite Element Analysis (composite unit cell) captures load transfer to the hydride phase
 - Short Fiber Composite Mechanics predicts critical size for load transfer and hydride fracture
 - Small beams enable mapping of hydride content and fracture around crack tips
- Related studies not discussed here
 - WAXS/SAXS during fracture of stainless-steel (Stubbins et al)

Outlook

APS renewal: 1-ID Upgrade

- Optimized undulator and customized hutches
- Expect >10x brilliance gain from undulator at 80keV
 - mostly horizontal → 2-d focusing
- Improved temporal resolution → kinetic studies
- Improved vertical resolution through R&D (thermal control, dedicated equipment)
 - ~100-500 nm theoretically possible with CRLs, with sufficiently low divergence for diffraction
- Reduction in $q(\text{min})$ for HE-SAXS
 - Through optimized detector (match potentially longer samp-det distances) and improved control of parasitic scattering (2-d focusing + slit system).
 - Access larger features and reduce ambiguity in analysis
- Increased use of complementary full-field imaging

APS → ERL

- Increased potential for 2-d focusing/temporal studies
- Increased coherence will enhance fidelity of imaging data

Acknowledgements

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 - More details at <http://me.queensu.ca/research/nuclear>
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